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Simulation Study for the Half-Life Measurement of ^{180m}Ta Using HPGe Detectors

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^{180m}Ta, which is the second excited state of ^{180}Ta at E_x = 77 keV, is naturally occurring and is the only known stable isomer. The half-life of ^{180m}Ta is considered to be an important parameter for nuclear synthesis models for heavy elements. However, the decay of ^{180m}Ta has never been observed even though several groups tried to measure it. We will search for gamma transitions from ^{180m}Ta decays in a tantalum sample by using an array of fourteen HPGe detectors recently installed in an underground laboratory at Yangyang, Korea. In preparation for the measurement, Monte-Carlo simulation studies were conducted to optimize the tantalum sample configuration. Based on the simulation study, we decided on a configuration composed of a 2 mm thick disk with diameter of 200 mm and six 2 mm thick rectangular plates with dimension of 158 × 195 mm^2. The finalized tantalum sample configuration gives 2.0 and 7.5 coincidence events per year for the EC and the β-decay of ^{180m}Ta, respectively. In this study, we used $T_{1/2} = 2.0 \times 10^{17}$ years and $T_{1/2} = 5.8 \times 10^{16}$ years for the EC and β-decay which are the present best lower-limits as reported by B. Lehnert *et al.*

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I. INTRODUCTION

^{180}Ta is the heaviest stable odd-odd nucleus and it naturally exists only in isomeric state, ^{180m}Ta [1,2]. ^{180m}Ta, a state with an excitation energy (E_x) of 77 keV and angular momentum and parity (J^π) of 9^-, is the rarest stable nuclide in nature with a natural abundance of 0.01201(8)% [3]. ^{180m}Ta is one piece of the puzzle for understanding nucleosynthesis of heavy elements because its production is bypassed in the general nucleosynthesis

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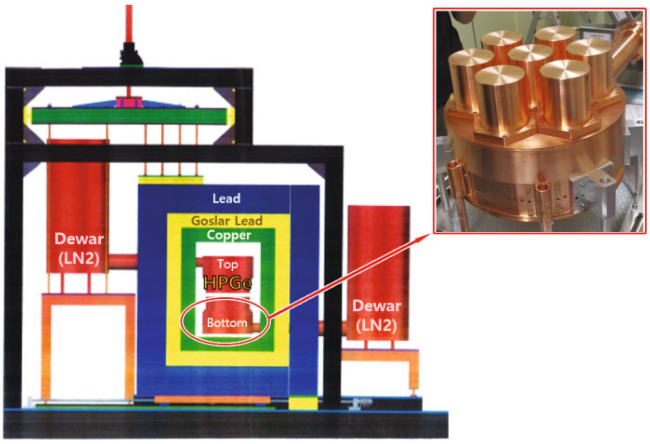

Fig. 1. (Color online) The structure of the CAGe system. The bottom and the top arrays consist of seven HPGe detectors each.

processes such as the r and the s processes [4,5].

Two expected decay modes of ^{180m}Ta are β-decay to ^{180}W and electron capture (EC) to ^{180}Hf. The branching ratios of these two decay modes from the isomeric state are critical for unknown inputs to isotope production processes. Even though the half-life of the ground state of ^{180}Ta is already known to be 8.154(6) hours [6], the half-life of ^{180m}Ta has never been measured [6–8]. The latest half-lives for the EC and the β-decay are $> 2.0 \times 10^{17}$ years and $> 5.8 \times 10^{16}$ years, respectively, and hence the total half-life of ^{180m}Ta is $> 4.5 \times 10^{16}$ years [9].

There were several studies for observing the decay of ^{180m}Ta using high-purity germanium (HPGe) detectors [10,11]. The center for underground physics (CUP) developed an array of fourteen HPGe detectors, which is called the CAGe (CUP array of high-purity germanium detectors). A tantalum sample measurement using the CAGe will be conducted to search for gamma transitions from the ^{180m}Ta decay. Because it is a very challenging experiment, Monte-Carlo studies are critical in designing the optimal experimental condition. In this paper, we describe the simulation studies for deciding the configuration of the tantalum sample. Furthermore, we estimate the count rate of gamma peaks from the ^{180m}Ta decay from the sample, with the best half-life limit.

II. EXPERIMENTAL APPARATUS

The CAGe was constructed at the Yangyang laboratory (Y2L) which is located in an underground tunnel with a depth of 700 m (2000 meter water equivalent) in Yangyang, Korea [12]. It was developed for high sensitivity measurements and rare decay experiments [13]. For the lifetime measurement of ^{180m}Ta, the CAGe has an advantage over a single HPGe detector because it is possible to identify coincidence events clearly.

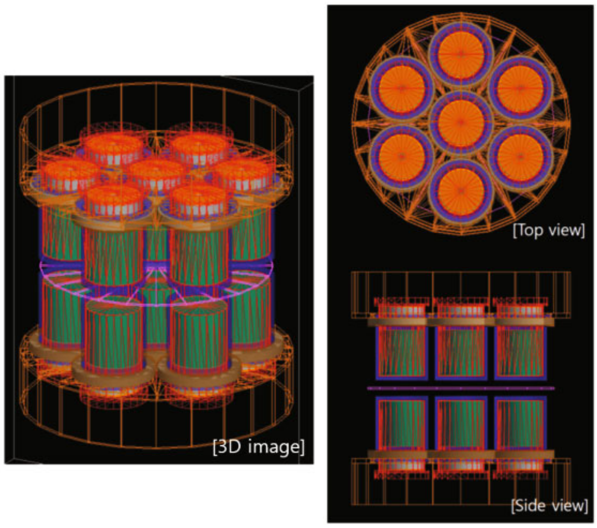

Fig. 2. (Color online) The geometry of the CAGe detectors.

The CAGe is composed of two horizontal arrays which are facing each other. Seven HPGe detectors are placed in each array as shown in Fig. 1. Each HPGe crystal has a 70% relative detection efficiency. These two arrays with detectors are called the 'top array' and the 'bottom array'. Samples can be placed in the middle of the two arrays for optimal efficiency, with extra material optionally placed around the outside of HPGe detectors. The distance between the top array and the bottom array can be adjusted from 2.5 cm to 7.5 cm. The HPGe detectors are shielded by lead and copper blocks to achieve an ultra-low background level. Thicknesses of the lead and the copper shields are 20 cm and 10 cm, respectively. The innermost lead shield consists of 5 cm thick Goslar lead, which has lower internal background than normal leads [14].

The configuration of a sample is an essential factor for the detection efficiency. Even though a larger sample volume is usually better than a smaller volume due to its higher statistics at a given time, the thickness of the tantalum sample for the ^{180m}Ta measurement has to be decided carefully because of the high density of the tantalum, 16.65 g/cm^3 [15]. We focus in coincidence events in this study. If we increase the thickness of the tantalum sample, coincidence events interacting with both the upper and lower arrays are suppressed. Therefore, there is an optimal thickness of the tantalum sample for this study.

Figure 2 shows a schematic drawing of the CAGe detectors using the GEANT4 tool [16]. There are two different types of space in the CAGe detectors for the sample installation; one is the central space between the top and the bottom arrays, and the other is the side space surrounding HPGe detectors. Three simulation studies were conducted to decide the most effective configuration of the tantalum sample.

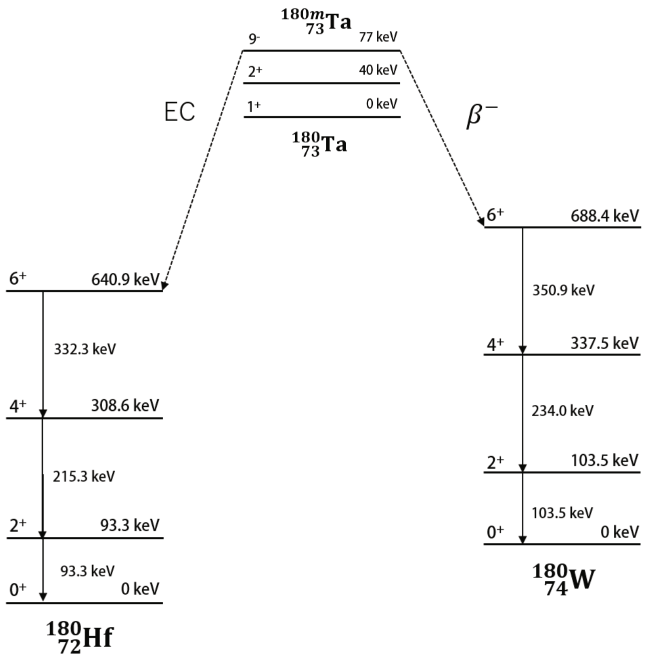

Fig. 3. Decay scheme of ^{180m}Ta.

III. SIMULATION AND ANALYSIS METHOD WITH GEANT4

1. Considerations for simulation studies

There are two decay modes of ^{180m}Ta, electron capture (EC) to ^{180}Hf and β-decay to ^{180}W, and each decay mode emits gamma rays of three different energies as shown in Fig. 3 [17]. However, these two decay modes were not observed to date. A clear signature for identifying either the EC mode or the β-decay mode of ^{180m}Ta is the detection of three sequential gamma transitions in coincidence. But the 93.3 keV and the 103.5 keV peaks, from the gamma transitions from the 2^+ states to the ground states of each decay mode, are difficult to observe because their transition probabilities are much lower than the other transitions, such as the $6^+ \rightarrow 4^+$ and $4^+ \rightarrow 2^+$ transitions [10], and their low energies are in the background continuum. Therefore, in our simulation studies, we neglected these two lowest energy peaks and only considered observation of the other four gamma peaks, which are the 350.9 keV and the 234.0 keV peaks in the EC and the 332.3 keV and the 215.3 keV peaks in the β-decay. The decay model in the GEANT4 tool (v.4.9.6) could not be used for this study because the tool does not contain the ^{180m}Ta decay in its database. For this reason, we generated gamma rays of two different energies corresponding to the sequential transitions for each decay mode. These two photons were generated at the same time, from the same position in the sample geometry, and with random direction vectors. One million pairs of two photons were generated within the sample geometry in each simulation.

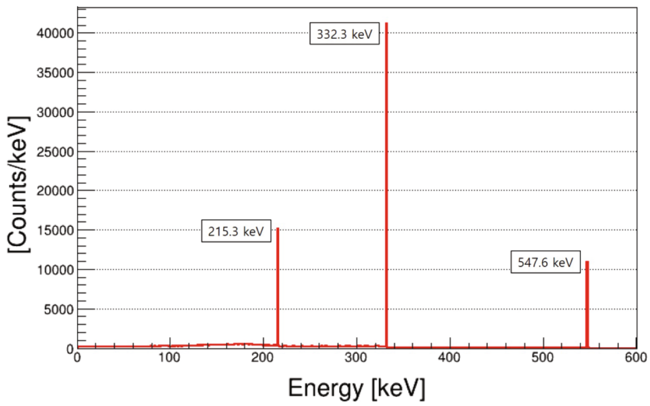

Fig. 4. (Color online) The energy spectrum of single-hit events from the simulation using one million primary particle pairs of two photons. The finalized sample geometry (Fig. 13) was used for this spectrum.

2. Analysis Method

We classify each detected event from the simulation as either a 'single-hit' event or a 'double-hit' event. Single-hit events are defined as those for which energy is only deposited in a single detector. This may be from a single photon, or from two photons interacting with a single detector. Double-hit events are defined as those for which energy is deposited in exactly two detector elements in coincidence. We ignore multiple-hit events involving more than two detectors because they are increasingly rare. According to our simulation results, double-hit events account for about 98% of all multiple-hit events.

The energy values of the gamma transitions for the EC and the β-decay are very similar to each other (Fig. 3). The difference between the gamma energies corresponding to the similar transition modes are less than 20 keV, and their detection efficiencies are in agreement within error, as shown in Fig. 5. Therefore, we describe our simulation studies for the EC mode only.

Figure 4 shows single-hit counts in the EC mode simulation when one million photon pairs were generated. Even though same number of photons were generated, the 332.3 keV peak contains more events than the 215.3 keV peak. A peak at 547.6 keV was observed when these two photons deposit their energies in one detector simultaneously.

The plot of double-hit events in the EC mode simulation is shown in Fig. 6. The x and y coordinates, E_1 and E_2 respectively, indicate the two energies deposited in two different HPGe detectors. We specified four regions in the plot, R1, R2, R3, and R4, to extract events of interest, specifically those which can be clearly identified with the decay by association with the transition energies. The events in the R1 region correspond to events where the full energies of the 332.3 keV and the 215.3 keV gamma rays are deposited separately in

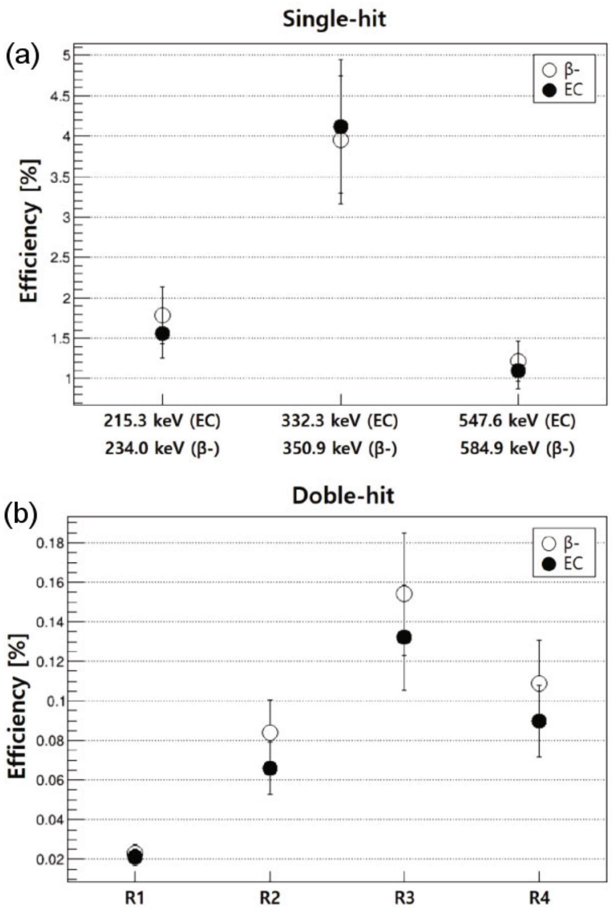

Fig. 5. The detection efficiencies for single-hit and double-hit events from the simulation with the finalized sample geometry. (a) Efficiency comparison for single-hit events and (b) Efficiency comparison for double-hit events.

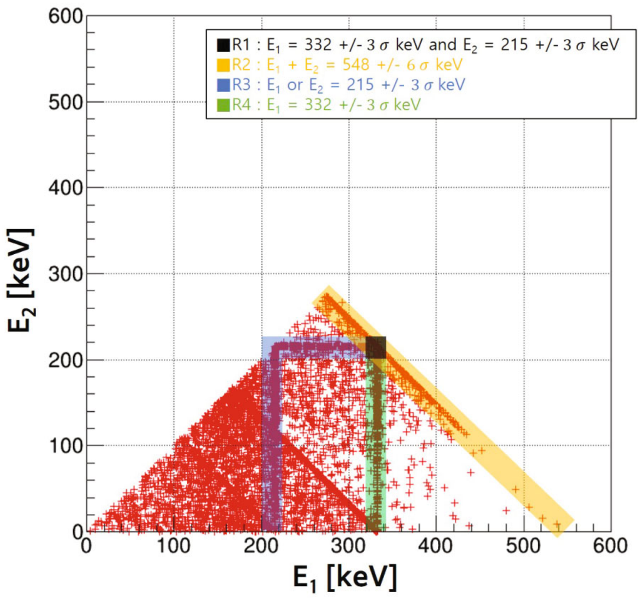

Fig. 6. (Color online) The plot of double-hit events from the simulation using the finalized sample geometry. E_1 and E_2 are the energies detected simultaneously in two different detector elements, with E_1 defined as being the higher of the two energies. The 1 σ energy resolution from the peak fitting is about 0.4 keV in this simulation study.

different detectors. This event class is the most specific in distinguishing real decays from backgrounds. The R2 region corresponds to events where the full energy of the two gammas was deposited in two detectors, but includes events where one or both gammas interacted with both detectors via Compton scattering. There are no peaks around 547.6 keV in background radioactivity in general [18]. Therefore, in the real tantalum measurement, after peak fitting and background continuum subtraction, double-hit events in the R2 region are associated with coincidence events from the 332.3 keV and the 215.3 keV gamma transitions. The R3 and the R4 regions correspond to events where one of the two gammas fully deposited in a single detector while the other gamma scattered in a separate detector by Compton scattering. We focused on the detection yield of the double-hit events more than the single-hit events, because the former have much lower background contributions. Hence, the purpose of this simulation study is to find the thickness and geometric configuration of a tantalum sample that gives the most double-hit events in the four regions.

IV. RESULT

Adjustments to the sample configuration for the Monte-Carlo simulation were separated into three categories: the thickness of the tantalum, the sample shape around the perimeter of the detector, and the diameter of the central sample disk. The primary particles generated were pairs of 215.3 keV and 332.3 keV gammas representing the first two gamma transitions in the EC mode from ^{180m}Ta.

1. Thickness selection

We conducted Monte-Carlo simulations to decide an effective thickness of a central sample, which will be placed between the top array and the bottom array. Considering the positioning of the CAGe detectors, a disk shape was considered for the central sample as shown in Fig. 7. The diameter of the central disk was fixed at 300 mm, which is the maximum value that can be placed in the CAGe.

The full energy peak efficiency(FEPE) from Monte-Carlo simulation can be calculated by $\varepsilon = \frac{N_{Det}}{N_{Gen}}$, where N_{Det} is the number of detected events in the peak of interest and N_{Gen} is the number of all the generated primary events [19]. Table 1 lists FEPEs from our simulation results. FEPE of the 215.3 keV peak is lower than that of the 332.3 keV peak because the lower energy gam-

Table 1. Detection efficiencies from simulations for the thickness selection.

(a) FEPE of single-hit events

Geometry ID	Thickness (mm)	Mass (kg)	FEPE (%) 215.3 keV	332.3 keV	547.6 keV
G1-1	1	1.18	3.37	6.81	2.76
G1-2	2	2.36	2.34	5.93	1.58
G1-3	3	3.54	1.71	5.03	1.0 7
G1-4	4	4.72	1.30	4.21	0.82
G1-5	5	5.90	1.05	3.58	0.66

(b) FEPE of double-hit events

Geometry ID	Thickness (mm)	Mass (kg)	FEPE (%) R1	R2	R3	R4
G1-1	1	1.18	0.035	0.138	0.312	0.192
G1-2	2	2.36	0.022	0.080	0.170	0.108
G1-3	3	3.54	0.011	0.045	0.110	0.067
G1-4	4	4.72	0.010	0.040	0.080	0.055
G1-5	5	5.90	0.007	0.030	0.070	0.041

mas are shielded more strongly by inactive layers in the detector crystals and surrounding materials. The estimated counts N can be obtained by

$$N = M \times t \times \varepsilon \times a, \quad (1)$$

where M is the mass of a tantalum sample, t is the measurement time, and ε is the FEPE. The specific activity, a, of the sample can be obtained by

$$a = \frac{\ln 2 \times N_A \times \mathrm{NA}}{T_{1/2} \times A_{r,Ta}}, \quad (2)$$

where $T_{1/2}$ is the half-life of ^{180m}Ta, $A_{r,Ta}$ is the atomic weight of tantalum, N_A is the Avogadro's number, and NA is the natural abundance of ^{180m}Ta. For our rate calculations we assumed a value for $T_{1/2}$ of 2.0×10^{17} years, which is the lower-limit according to the recent experiment [9]. The calculated value of a is about 1.38×10^3 /kg/yr using Eq. (2).

Figure 8 compares the estimated peak rates from the simulation. In this study, we concentrate on the CAGe's advantages by considering double-hit events. The estimated numbers of double-hit events from five geometries were no difference within the error. By considering the single-hit events and the double-hit events together, we decided to choose 2 mm for the thickness of the central disk.

2. Configuration selection

As shown in Fig. 9, there are different ways of adding more tantalum samples vertically in the inner space between the HPGe detectors and in the outer space surrounding all fourteen HPGe detectors. These vertical samples can increase the detection yield compared to the central disk sample only. Our second Monte-Carlo simulations were conducted to decide an efficient geometry of the vertical sample that surrounds the HPGe detectors. All G2 geometries include the G1-2 geometry, which is the central disk sample with 2 mm thickness and 300 mm diameter. Thicknesses of vertical samples were fixed at 2 mm based on the first simulation results for the thickness selection.

Table 2 lists the FEPE results from simulations, and Fig. 10 shows the estimated rates of single-hit and double-hit events using Eq. (1) and Eq. (2). According to calculation results, there is a distinct difference in this simulation depending on the configuration of the vertical sample. Among the candidate geometries considered, the G2-2 geometry is the best for optimizing double-hit events.

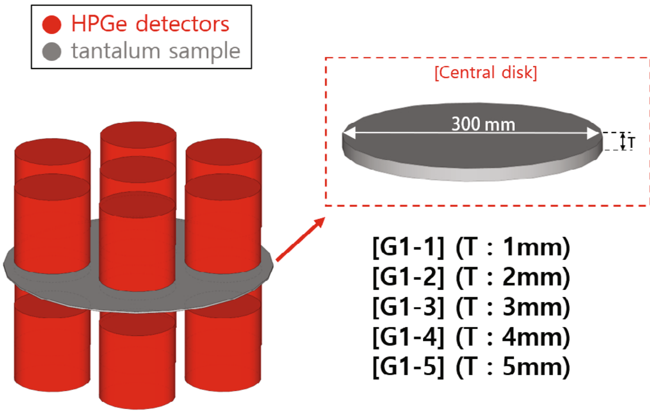

Fig. 7. (Color online) Geometries of the samples and detectors for determining the thickness of the central disk sample.

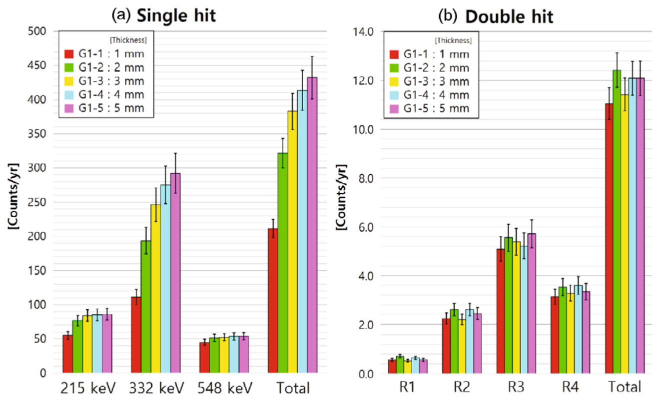

Fig. 8. (Color online) Estimated ^{180m}Ta count rates for different central disk thicknesses. Rates are shown for the single-hit gamma peaks, and for our four double-hit coincidence requirements. $T_{1/2}$ for the calculation was equal to the present limit of 2.0×10^{17} years [9]. (a) Single-hit counts per year and (b) double-hit counts per year.

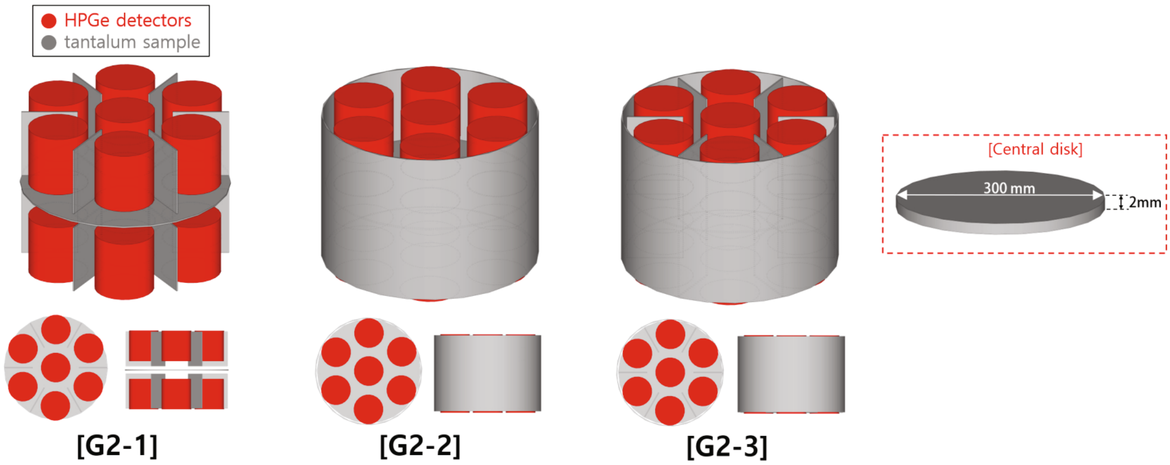

Fig. 9. (Color online) Geometries of the samples and detectors for determining the vertical sample shape.

Table 2. Detection efficiencies from simulations for the configuration selection.

(a) FEPE of single-hit events

Geometry ID	Mass (kg)	FEPE (%) 215.3 keV	332.3 keV	547.6 keV
G2-1	5.56	1.94	5.06	1.36
G2-2	8.21	1.47	3.81	1.02
G2-3	11.42	1.44	3.76	1.00

(b) FEPE of double-hit events

Geometry ID	Mass (kg)	FEPE (%) R1	R2	R3	R4
G2-1	5.56	0.009	0.031	0.063	0.041
G2-2	8.21	0.014	0.049	0.096	0.062
G2-3	11.42	0.009	0.022	0.041	0.030

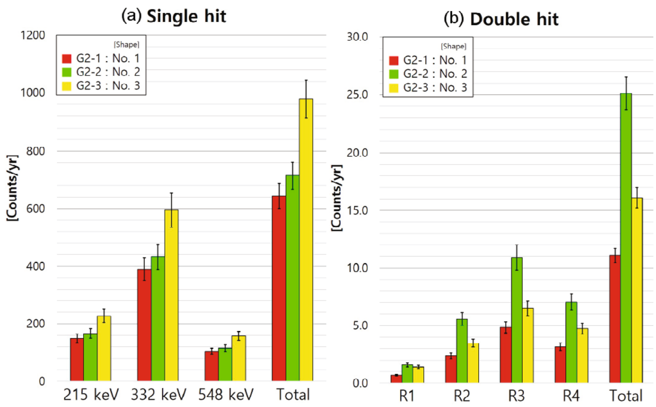

Fig. 10. (Color online) Estimated ^{180m}Ta count rates from the simulations using different geometries for the vertically oriented sample pieces, assuming $T_{1/2}$ is equal to the present limit of 2.0 $\times$ 10^{17} years [9]. (a) Single-hit counts per year and (b) double-hit counts per year.

3. Diameter selection

Of the configurations considered, the G2-2 sample geometry, which has the cylindrical vertical sample and the central disk sample, was identified as the most effective for double-hit detection efficiency. We considered the central disk again to find the most effective diameter without changing any other parameters in the G2-2 geometry. As shown in Fig. 11, the vertical sample geometry for G3 simulations was the same cylindrical tube shape as used in the previous G2 study. There were three simulations for three different diameters of the central disk and one simulation without the central disk.

Table 3 lists simulated FEPEs and Fig. 12 shows estimated counts per year from simulations. The estimated counts of double-hit events in this study increases, as the diameter of the central disk sample increases except for simulations using the G3-4 geometry. The reason for this may be due to the fact that the central disk sample with 300 mm diameter covered most of the area between the top and the bottom arrays and it acts as a shield for coincidence events from the vertical samples. The best configuration for having both single-hit events and double-hit events while minimizing the sample materials due to intrinsic background was the G3-3 geometry.

4. Final Configuration

After all our simulations, the G3-3 geometry, consisting of a 2 mm thick and 200 mm diameter central disk and 2 mm thick vertical sample, was selected because it gives the largest double-hit event rate of all the geometries. Considering the convenience of the manufacturing process, the cylindrical tube was replaced by hexagonal panels for the vertical sample as shown in Fig. 13. The configuration is composed of one central disk and six vertical panels resulting in a total mass of about 6.9 kg. Six

Table 3. Detection efficiencies from simulations for the diameter selection.

(a) FEPE of single-hit events

Geometry ID	Diameter (mm)	Mass (kg)	FEPE (%) 215.3 keV	332.3 keV	547.6 keV
G3-1	No disk	5.86	1.31	3.41	0.89
G3-2	100	6.12	1.36	3.58	0.94
G3-3	200	6.90	1.50	3.84	1.01
G3-4(G2-2)	300	8.21	1.47	3.81	1.02

(b) FEPE of-double hit events

Geometry ID	Diameter (mm)	Mass (kg)	FEPE (%) R1	R2	R3	R4
G3-1	No disk	5.86	0.022	0.069	0.134	0.090
G3-2	100	6.12	0.022	0.066	0.142	0.092
G3-3	200	6.90	0.020	0.062	0.128	0.084
G3-4(G2-2)	300	8.21	0.014	0.049	0.096	0.062

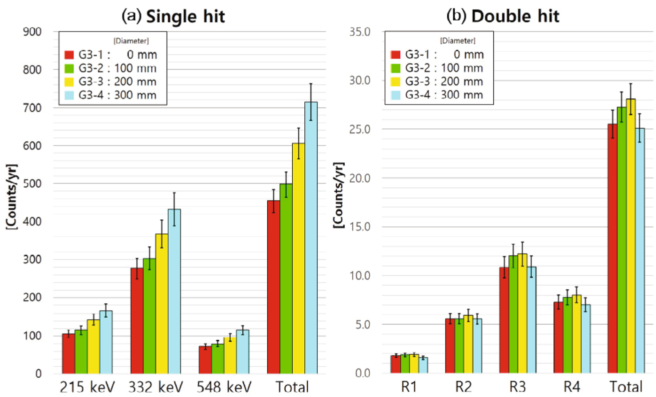

Fig. 12. (Color online) Estimated ^{180m}Ta count rates for different diameters of the central disk assuming $T_{1/2}$ is equal to the present limit of 2.0 × 10^{17} years [9]. All geometries in this simulation study contain the same tube-geometry for the vertical sample, together with the central disk sample. (a) Single-hit counts per year and (b) Double-hit counts per year.

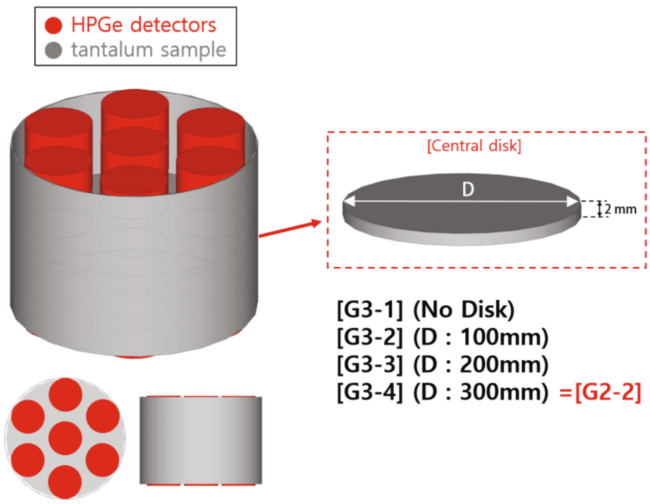

Fig. 11. (Color online) Geometries of the samples and detectors for determining the diameter of the central disk sample.

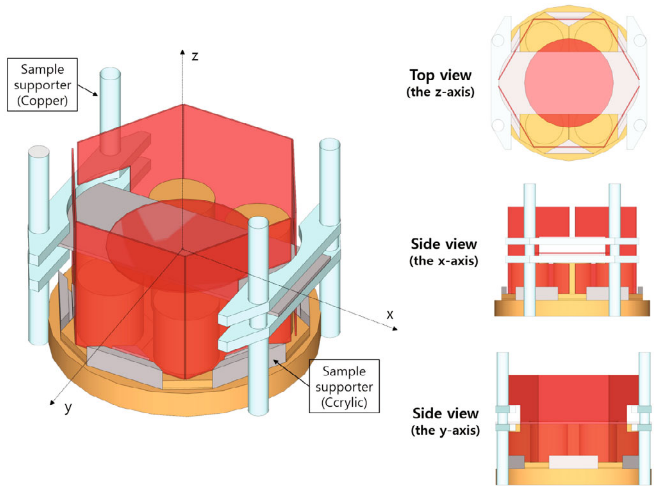

Fig. 13. (Color online) Final sample configuration including the CAGe detectors and sample supporters. The structure (in red) is the finalized tantalum sample configuration.

panels of tantalum were used to cover the outer part of the CAGe detectors in a way similar to the tube configuration used in the simulation. The dimension size of each panel is 158 × 195 mm^2. Using this final configuration and the stated half-life assumption, the estimated rates of single-hit events for 215.3 keV, 332.3 keV, and 547.6 keV are 149, 392, and 105 per year, respectively. The estimated rates of double-hit events in the R1, the R2, the R3, and the R4 regions for the EC are 2.0, 6.3, 12.6, and 8.6 per year, respectively. These estimated rates are nearly the same as the results obtained when using the G3-3 geometry. We also calculated the count rate for the β-decay using the simulation result with this final configuration. The $T_{1/2}$ was assumed equal to the present limit of 5.8 × 10^{16} years [9]. The estimated rates of single-hit events for 234.0 keV, 350.9 keV, and 584.9 keV are 584, 1298, and 400, respectively. The estimated rates of double-hit events in the R1, the R2, the R3, and the R4 regions for the β-decay are 7.5, 27.6, 50.5, and 35.8 per year, respectively.

V. CONCLUSION

We plan to measure a tantalum sample using CAGe to observe the rare decay of ^{180m}Ta and find its half-life. Monte Carlo simulations using the GEANT4 tool were conducted to identify the geometric configuration of the sample which optimizes double-hit coincidence detection. Simulations were carried out in order to consider the optimal thickness of the central disk sample, the optimal configuration of the vertical sample geometry, and finally within that configuration, the optimal diameter for the central disk sample. From our simulation results, the thickness and the diameter for the central disk sample were determined as 2 mm and 200 mm, respectively. For the vertical sample, the hexagonal geometry was chosen considering simulation result and manufacturing constraints. Figure 13 shows the final sample configuration, which is composed of one central disk sample and a hexagonal vertical sample. The vertical sample was constructed with six vertical panels with dimension of 158 × 195 mm^2.

We estimated the peak rates from the finalized tantalum sample configuration, assuming $T_{1/2}$ equals to 2.0×10^{17} years and 5.8×10^{16} years, the best lower-limit values for the EC and β-decay, respectively [9]. For the EC, the estimated rates of single-hit events for the 215.3 keV, the 332.3 keV and the 547.6 keV peaks are 149, 392, and 105 per year, respectively. The estimated rates of double-hit events in the R1, the R2, the R3, and the R4 regions for the EC are 2.0, 6.3, 12.6, and 8.6 per year, respectively. For the β-decay, the estimated rates of single-hit events for 234.0 keV, 350.9 keV, and 584.9 keV peaks are 584, 1298, and 400, respectively. The estimated rates of double-hit events in the R1, the R2, the R3, and the R4 regions for the β-decay are 7.5, 27.6, 50.5, and 35.8 per year, respectively. Based on this study, the tantalum samples of the final configuration were prepared and installed in the CAGe detectors for measuring the half-life of ^{180m}Ta.

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